Int'l Application No.: PCT/GB2004/002859 Docket No.: 31229-226445

AMENDMENTS TO THE CLAIMS

Claim 1. (Currently Amended) A process for the hydroformylation of ethylenically unsaturated compounds, which process comprises reacting said ethylenically unsaturated compound with carbon monoxide and hydrogen, in the presence of a catalyst system, the catalyst system obtainable by combining:

- a) a Group VIII metal compound; and
- b) a bidentate compound of general formula (Ia)

(Ia)

wherein:

R is a bridging group[[;]] selected from the group consisting of:

an alkylene group; and

A-Ar-B, in which Ar is an optionally substituted aryl moiety to which A and B are linked on available adjacent carbon atoms, and A and B each independently represent lower alkylene;

R¹ to R¹² each independently represent lower alkyl, aryl or Het;

Q1 and Q2 each independently represent phosphorus, arsenic or antimony,

the process characterised in that a chlorine moiety is present in at least said Group VIII metal compound.

Claim 2. (Currently Amended) A process for the hydroformylation of ethylenically unsaturated compounds, which process comprises reacting said ethylenically unsaturated compound with carbon monoxide and hydrogen, in the presence of a catalyst system and a solvent, the catalyst system obtainable by combining:

- a) a metal of Group VIII or a compound thereof; and
- b) a bidentate compound of general formula (Ia)

(Ia)

wherein:

R is a bridging group[[;]] selected from the group consisting of: an alkylene group; and

A-Ar-B, in which Ar is an optionally substituted aryl moiety to which A and B are linked on available adjacent carbon atoms, and A and B each independently represent lower alkylene;

R¹ to R¹² each independently represent lower alkyl, aryl or Het;

 Q^1 and Q^2 each independently represent phosphorus, arsenic or antimony,

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the process characterised in that a chlorine moiety is present in at least one of the said Group VIII metal compound or said solvent.

- Claim 3. (Previously presented) The process as claimed in claim 2, wherein the chlorine moiety is present in both said Group VIII metal compound and said solvent.
- Claim 4. (Previously Presented) The process as claimed in claim 1, wherein R^1 to R^{12} each independently represent C_1 to C_6 alkyl, C_1 to C_6 alkyl phenyl or phenyl.
- Claim 5. (Original) The process as claimed in claim 4, wherein R^1 to R^{12} each independently represent methyl, ethyl, n-propyl, iso-propyl, n-butyl, iso-butyl, tert-butyl, pentyl, hexyl and cyclohexyl.
- Claim 6. (Previously Presented) The process as claimed in claim 1, wherein R^1 , R^4 , R^7 and R^{10} each independently represent the same C_1 - C_6 alkyl; R^2 , R^5 , R^8 and R^{11} , each independently represent the same C_{1-6} alkyl; and R^3 , R^6 , R^9 and R^{12} , each independently represent the same C_{1-6} alkyl.
- Claim 7. (Previously Presented) The process as claimed in claim 1, wherein R^1 to R^{12} each represents the same C_1 - C_6 alkyl group.
- Claim 8. (Original) The process as claimed in claim 7, wherein the said C_{1-6} alkyl group is non-substituted and selected from the list comprising: methyl, ethyl, n-propyl, iso-propyl, n-butyl, iso-butyl, tert-butyl, pentyl, hexyl and cyclohexyl.
- Claim 9. (Original) The process as claimed in claim 8, wherein the said C_{1-6} alkyl group is methyl.

Claim 10. (Previously Presented) The process as claimed in claim 1, wherein Q^1 and Q^2 represents phosphorus.

Claim 11. (Previously Presented) The process as claimed in claim 1 wherein R is defined as -A-(K,D)Ar(E,Z)-B- and the bidentate compound is of general formula (I)

(I)

wherein:

Ar is a bridging group comprising an optionally substituted aryl moiety to which A and B are linked on available adjacent carbon atoms;

A and B each independently represent lower alkylene;

K, D, E and Z are substituents of the aryl moiety (Ar) and each independently represent hydrogen, lower alkyl, aryl, Het, halo, cyano, nitro, OR^{19} , $OC(O)R^{20}$, $C(O)R^{21}$, $C(O)OR^{22}$, $NR^{23}R^{24}$, $C(O)NR^{25}R^{26}$, $C(S)R^{25}R^{26}$, SR^{27} , $C(O)SR^{27}$, or -J-Q³($CR^{13}(R^{14})(R^{15})$) $CR^{16}(R^{17})(R^{18})$ where J represents lower alkylene; or two adjacent groups selected from K, Z, D and E together with the carbon atoms of the aryl ring to which they are attached form a further phenyl ring, which is optionally substituted by one or more substituents selected from hydrogen, lower alkyl, halo, cyano,

nitro, OR^{19} , $OC(O)R^{20}$, $C(O)R^{21}$, $C(O)OR^{22}$, $NR^{23}R^{24}$, $C(O)NR^{25}R^{26}$, $C(S)R^{25}R^{26}$, SR^{27} or $C(O)SR^{27}$;

R¹ to R¹⁸ each independently represent lower alkyl, aryl, or Het;

R¹⁹ to R²⁷ each independently represent hydrogen, lower alkyl, aryl or Het;

Q¹, Q² and Q³ (when present) each independently represent phosphorous, arsenic or antimony.

Claim 12. (Original) The process as claimed in claim 11, wherein R^1 to R^{18} each independently represent C_1 to C_6 alkyl, C_1 to C_6 alkyl phenyl or phenyl.

Claim 13. (Original) The process as claimed in claim 12, wherein R¹ to R¹⁸ each independently represent methyl, ethyl, n-propyl, iso-propyl, n-butyl, iso-butyl, tert-butyl, pentyl, hexyl and cyclohexyl.

Claim 14. (Previously Presented) The process as claimed in claim 11 wherein R^1 , R^4 , R^7 , R^{10} , R^{13} and R^{16} each independently represent the same C_1 - C_6 alkyl; R^2 , R^5 , R^8 , R^{11} , R^{14} and R^{17} each independently represent the same C_{1-6} alkyl; and R^3 , R^6 , R^9 , R^{12} , R^{15} and R^{18} each independently represent the same C_{1-6} alkyl.

Claim 15. (Previously Presented) The process as claimed in claim 11, wherein R^1 to R^{18} each represents the same C_1 - C_6 alkyl group.

Claim 16. (Original) The process as claimed in claim 15, wherein the said C_{1-6} alkyl group is non-substituted and selected from the list comprising: methyl, ethyl, n-propyl, iso-propyl, n-butyl, iso-butyl, tert-butyl, pentyl, hexyl and cyclohexyl.

Claim 17. (Original) The process as claimed in claim 16, wherein the said C_{1-6} alkyl group is methyl.

Claim 18. (Previously Presented) The process as claimed in claim 11, wherein Q^1 , Q^2 and Q^3 (when present) represents phosphorus.

Claim 19. (Previously Presented) The process as claimed in claim 11, wherein A, B and J (when present) each independently represent C₁ to C₆ alkylene.

Claim 20. (Original) The process as claimed in claim 19, wherein each of A, B and J (when present) represent –CH₂-.

Claim 21. (Previously Presented) The process as claimed in claim 11, wherein K, D, E and Z each represent hydrogen, phenyl, C_1 - C_6 alkylphenyl or C_1 - C_6 alkyl.

Claim 22. (Original) The process as claimed in claim 21, wherein K, D, E and Z each represent hydrogen.

Claim 23. (Original) The process as claimed in claim 11 wherein in formula (I):

A and B each independently represent unsubstituted C₁ to C₆ alkylene;

K, D, Z and E each independently represent hydrogen, C_1 - C_6 alkyl, phenyl, C_1 - C_6 alkylphenyl or -J- $Q^3(CR^{13}(R^{14})(R^{15}))CR^{16}(R^{17})(R^{18})$ where J represents unsubstituted C_1 to C_6 alkylene; or two of K, D, Z and E together with the carbon atoms of the aryl ring to which they are attached form a phenyl ring which is optionally substituted by one or more substituents selected from lower alkyl, phenyl or lower alkylphenyl;

 R^1 to R^{18} each independently represent C_1 to C_6 alkyl, phenyl or C_1 to C_6 alkylphenyl.

Claim 24. (Original) The process as claimed in claim 11 wherein in formula (I):

A and B both represent $-CH_2$ - or C_2H_4 ;

K, D, Z and E each independently represent hydrogen, C_1 - C_6 alkyl phenyl or C_1 - C_6 alkyl or -J- $Q^3(CR^{13}(R^{14})(R^{15}))CR^{16}(R^{17})(R^{18})$ where J is the same as A; or two of K, D, E and Z together with the carbon atoms of the aryl ring to which they are attached form an unsubstituted phenyl ring;

 R^1 to R^{18} each independently represent C_1 to C_6 alkyl;

Claim 25. (Original) The process as claimed in claim 24, wherein A and B both represent -CH₂-.

Claim 26. (Original) The process as claimed in claim 11, wherein in formula (I): each R^1 to R^{12} is the same and represents methyl;

A and B are the same and represent -CH₂-;

K, D, Z and E are the same and represent hydrogen.

Claim 27. (Previously Presented) The process as claimed in claim 11, wherein the reaction is carried out at a temperature of between 20°C and 180°C.

Claim 28. (Original) The process as claimed in claim 27, wherein the temperature is in the range 50°C to 150°C.

Claim 29. (Previously Presented) The process as claimed in claim 11, wherein the reaction is carried out under a partial pressure of carbon monoxide/hydrogen in the range of 1 to 700 bar.

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Claim 30. (Original) The process as claimed in claim 29, wherein the partial pressure is in the range 1 to 300 bar.

- Claim 31. (Previously Presented) The process as claimed in claim 11, wherein said ethylenically unsaturated compound has 1 to 3 carbon-carbon double bonds per molecule.
- Claim 32. (Original) The process as claimed in claim 31, wherein said compound has 1 carbon-carbon double bond per molecule.
- Claim 33. (Previously Presented) The process as claimed in claim 11, wherein the amount of bidentate compound of formula (I) to unsaturated compound is in the range 10⁻⁵ to 10⁻² moles per mole of unsaturated compound.
- Claim 34. (Previously Presented) The process as claimed in claim 11, wherein said catalyst system further comprises a support.
- Claim 35. (Previously Presented) The process as claimed in claim 11, wherein said bidentate compound is a phosphine selected from the group comprising bis (di-t-butyl phosphino)-o-xylene; 1,2 bis (diadamantylphosphinomethyl) benzene; 1,2 bis (diadamantylphosphinomethyl) naphthalene; 1,2 bis (di-t-pentyl phosphino)-o-xylene; and bis 1,2 (di-t-butyl phosphino) naphthalene.
- Claim 36. (Previously Presented) The process as claimed in claim 11, wherein the mol:mol range of compounds of formula (I) to Group VIII metal is in the range of 1:1 to 3:1.
- Claim 37. (Original) The process as claimed in claim 36, wherein said mol:mol range is in the range of 1:1 to 1.25:1.

Claim 38. (Previously Presented) The process as claimed in claim 11, wherein the Group VIII metal is selected from the group: cobalt, nickel, palladium, rhodium, ruthenium and platinum.

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Claim 39. (Original) The process as claimed in claim 38, wherein said Group VIII metal is rhodium.

Claim 40. (Previously Presented) The process as claimed in claim 11, wherein said chlorine moiety is present in at least said Group VIII metal compound and said compound is selected from the group comprising: [RhCl(CO)₂]₂, [RhCl(Cod)₂]₂, RhCl₃.xH₂O, [RhCl(Norbornadiene)]₂, [RhCl(Cyclooctene)₂]₂, Chloro(1,5-hexadiene)-rhodium(I)dimer, μ -dichlorotetraethylene-dirhodium, (bicyclo[2,2,1]hepta-2-5-diene)chlororhodium(I)dimer.

Claim 41. (Previously Presented) The process as claimed in claim 11, wherein said chlorine moiety is present in at least said Group VIII metal compound and said compound is selected from the group comprising: [RhCl(CO)₂]₂, [RhCl(Cod)₂]₂ and RhCl₃.xH₂O.

Claim 42. (Previously Presented) The process as claimed in claim 11, wherein said chlorine moiety is present in at least said solvent and said solvent is selected from the group consisting of dichloromethane, chlorobenzene, o-dichlorobenzene, m-chlorobenzene, carbon tetrachloride, trichloroethanes, dichloroethanes, chlorofluorocarbons (CFC's), tetrachloroethanes and tetrachloroethene.

Claim 43. (Original) The process as claimed in claim 42, wherein said solvent is dichloromethane.

Claim 44. (Previously Presented) The process as claimed in claim 11, wherein said ethylenically unsaturated compound has 2 to 20 carbon atoms.

Claim 45. (Original) The process as claimed in claim 44, wherein said compound has 5 to 15 carbon atoms.

Claim 46. (Original) The process as claimed in claim 44, wherein said compound has 6 to 12 carbon atoms.

Claim 47. (Previously Presented) The process as claimed in claim 11, wherein said ethylenically unsaturated compound is selected from the group comprising ethene, propene, 1-butene, 2-butene, isobutene, 1-pentene, 2-pentene, 3-pentene and branched isomers thereof, 1-hexene and its isomers, 1-heptene and its isomers, 1-octene and its isomers, 1-nonene and its isomers, 1-decene and its isomers, the C₁₁-C₂₀ alkenes and their known isomers, 3-pentenenitrile, methyl-3-penteneoate, 1,3 butadiene, 1,3-pentadiene, 1,3 hexadiene, 1,3 cyclohexadiene, 2,4-leptadiene, and 2-methyl 1,3 butadiene.

Claim 48. (Previously Presented) The process as claimed in claim 11, wherein the said catalyst system further comprises a polymeric dispersant dissolved in a liquid carrier, said polymeric dispersant being capable of stabilising a colloidal suspension of particles of the Group VIII metal or metal compound of the catalyst system within the said liquid carrier.

Claim 49. (Original) The process as claimed in claim 48, wherein said polymeric dispersant is selected from the list comprising: polyvinylpyrrolidone, polyacrylamide, polyacrylonitrile, polyethylenimine, polyglycine, polyacrylic acid, polymethacrylic acid, poly(3-hydroxybutyricacid), poly-L-leucine, poly-L-methionine, poly-L-proline, poly-L-serine, poly-L-tyrosine, poly(vinylbenzenesulphonic acid) and poly(vinylsulphonic acid).

Claim 50. (Original) The process as claimed in claim 49, wherein said polymeric dispersant is selected from the list comprising: polyvinylpyrrolidone and polyacrylic acid.

Claim 51. (Previously Presented) The process as claimed in claim 11, wherein the solvent is formed by one or more of the reactants, products or by-products of the process rather than being a separate entity.

Claim 52. (Previously Presented) A process as claimed in claim 11, wherein Ar is defined as including six-to-ten-membered carbocyclic aromatic groups, which groups are optionally substituted with, in addition to K, D, E or Z, one or more substituents selected from aryl, lower alkyl, Het, halo, cyano, nitro, OR^{19} , $OC(O)R^{20}$, $C(O)R^{21}$, $C(O)OR^{22}$, $NR^{23}R^{24}$, $C(O)NR^{25}R^{26}$, SR^{27} , $C(O)SR^{27}$ or $C(S)NR^{25}R^{26}$ wherein R^{19} to R^{27} each independently represent hydrogen, aryl or lower alkyl.

Claim 53. (Previously Presented) A process as claimed in claim 1 wherein at least one (CR^xR^yR^z) group attached to Q¹ and/or Q², i.e. CR¹R²R³, CR⁴R⁵R⁶, CR⁷R⁸R⁹, or CR¹⁰R¹¹R¹², may instead be congressyl or adamantyl, or both groups defined above as (CR^xR^yR^z) attached to either or both Q¹ and/or Q², may, together with either Q¹ or Q² as appropriate, instead form an optionally substituted 2-phospha-tricyclo[3.3.1.1{3,7}]decyl group or derivative thereof.

Claim 54. (Previously Presented) A process as claimed in claim 11, wherein Ar is a cyclopentadienyl group, and Z may be represented by $-M(L_1)_n(L_2)_m$ and Z is connected via a metal ligand bond to the cyclopentadienyl group, M represents a Group VIB or VIIIB metal or metal cation thereof; and

 L_1 represents a cyclopentadienyl, indenyl or aryl group each of which groups are optionally substituted by one or more substituents selected from hydrogen, lower alkyl, halo, cyano, nitro, OR^{19} , $OC(O)R^{20}$, $C(O)R^{21}$, $C(O)OR^{22}$, $NR^{23}R^{24}$, $C(O)NR^{25}R^{26}$, $C(S)R^{25}R^{26}$, SR^{27} , $C(O)SR^{27}$ or ferrocenyl;

 L_2 represents one or more ligands each of which are independently selected from hydrogen, lower alkyl, alkylaryl, halo, CO, $PR^{43}R^{44}R^{45}$ or $NR^{46}R^{47}R^{48}$;

R⁴³ to R⁴⁸ each independently represent hydrogen, lower alkyl, aryl or Het;

n = 0 or 1;

and m = 0 to 5;

provided that when n = 1 then m equals 0, and when n equals 0 then m does not equal 0.

Claim 55. (Previously Presented) A process as claimed in claim 1, wherein R represents an alkylene bridging group.

Claim 56. (Canceled).

Claim 57. (Canceled).

Claim 58. (Currently Amended) A hydroformylation reaction catalyst system for the catalysis of ethylenically unsaturated compounds with carbon monoxide and hydrogen in the presence of said system, the catalyst system obtainable by combining:

- a) a Group VIII metal compound as defined in claim 1; and
- b) a bidentate compound as defined in claim 1, wherein the R bridging group is
 - i) an alkylene selected from the group consisting of ethylene and substituted variants thereof, propylene and substituted variants thereof, and butylene and substituted variants thereof; or
- ii) A-Ar-B, in which Ar is an optionally substituted aryl moiety to which A and B are linked on available adjacent carbon atoms, and A and B each independently represent lower alkylene;
 and wherein the catalyst system is characterised in that a chlorine moiety is present in at least said
 Group VIII metal compound.

Claim 59. (Canceled).

Claim 60. (Previously Presented) A hydroformylation reaction medium, wherein said medium comprises a catalyst system and a solvent, wherein said system is for use in a process for the hydroformylation of ethylenically unsaturated compounds and which process comprises reacting said ethylenically unsaturated compound with carbon monoxide and hydrogen, in the presence of said system and said solvent, wherein said solvent is as defined in claim 2, the catalyst system obtainable by combining:

- a) a metal of Group VIII or a compound thereof as defined in claim 1; and
- b) a bidentate compound as defined in claim 1,

and wherein the reaction medium is characterised in that a chlorine moiety is present in at least one of the said Group VIII metal compound or said solvent, including the possibility of being present in both.

Claim 61. (Previously Presented) A process for the hydroformylation of ethylenically unsaturated compounds, said process comprising the step of reacting said ethylenically unsaturated compound with carbon monoxide and hydrogen, in the presence of a catalyst system as defined in claim 1.

Claim 62. (Previously Presented) A process for the hydroformylation of ethylenically unsaturated compounds, said process comprising the step of reacting said ethylenically unsaturated compound with carbon monoxide and hydrogen, in the presence of a reaction medium as defined in claim 60.

Claim 63. (Previously Presented) The process of claim 55, wherein R represents a lower alkylene bridging group.

Claim 64 (New): The process of claim 1, wherein R is a lower alkylene or A-Ar-B, in which Ar is an optionally substituted aryl moiety to which A and B are linked on available adjacent carbon atoms, and A and B each independently represent lower alkylene.

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Claim 65 (New): The process of claim 64, wherein the lower alkylene is selected from the group consisting of methylene and substituted variants thereof, ethylene and substituted variants thereof, propylene and substituted variants thereof, and butylene and substituted variants thereof.